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TITLE: MEASUREMENTS OF INDUCED RADIOACTIVITY IN SOME LDEF SAMPLES

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## MEASUREMENTS OF INDUCED RADIOACTIVITY IN SOME LDEF SAMPLES

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### SUMMARY

Twenty-six stainless steel trunnion samples, five aluminum end support retainer plate samples, two aluminum keel plate samples, and two titanium clips were analyzed. The shielded high-purity germanium detectors used had efficiencies of 33%, 54%, and 80% at 1332 keV. Detector efficiencies as a function of energy and corrections for self-absorption in the samples were determined with calibrated sources and unactivated control samples. Several measurements were made on most samples. In the trunnion samples,  $^{54}\text{Mn}$  and  $^{57}\text{Co}$  were seen and limits were obtained for other isotopes. The results agree well with 1-dimensional activation calculations for an anisotropic trapped proton model. In the aluminum and titanium samples,  $^{22}\text{Na}$  was seen. Other results are presented.

### INTRODUCTION

Los Alamos National Laboratory is one of several laboratories involved in the analysis of induced radioactivity in samples from the Long Duration Exposure Facility (LDEF). We analyzed samples of the spacecraft rather than samples deliberately placed on board. The goal of this work was to provide data that could be used in modeling calculations to determine the integrated radiation environment at the LDEF. From these results the radiation environment of Space Station Freedom and other spacecraft in similar low-Earth orbits can be predicted.

The 20 samples from section D of the trunnions were received about 65 days after the shuttle Columbia landed with LDEF at 12:30 am on 20 January 1990. We corrected all induced radioactivities to this time. The end support retainer plate, trunnion sections C, H, and N, titanium clip, and keel plate samples were received about 155, 178, 200, and 430 days, respectively, after this time.

## EXPERIMENTAL DETAILS

### Detectors

We used three high purity germanium detectors to measure gamma rays from the samples. The smallest two had efficiencies of 33% and 54% relative to a 3" x 3" NaI(Tl) scintillator. Each was part of an automated counting system (figure 1) used for programmatic work at Los Alamos. The germanium crystal was shielded from background by several inches of lead. The samples were mounted on thin aluminum plates and placed on the carousel. As each station came into position under the detector, a hydraulic ram pushed the sample and sample holder up into position such that the aluminum plate was about 1.46 cm from the detector. Data were acquired into a multichannel analyzer and transferred to a computer for storage and analysis. The spectra, which had 4096 channels from ~50 keV to 2 MeV, were analyzed with two codes based on the GAMANAL spectral analysis code of Gunnink at Lawrence Livermore National Laboratory. (ref. 1).

The largest detector (figure 2) had an efficiency of 80%. It was mounted on a portable LN2 dewar for field nuclear safeguards work. The sample was placed in contact with the detector can, which was shielded by 2 to 4 inches of lead. The data were acquired in a PC-based multichannel analyzer operated manually. Peak areas in the spectra were determined with the code MAESTRO from EG&G ORTEC.

### Efficiencies

The detector efficiencies were determined in geometries similar to those used to measure the samples. Each sample had to be placed close to the detector, which meant that the efficiencies were very sensitive to the distance from the detector. Table I lists the radioactive nuclides, half-lives, and gamma-ray energies in the mixed calibration source provided by A. Harmon of the Marshall Space Flight Center. The activity was contained in many small spots placed in a matrix on a 2" x 2" sheet of mylar 0.002 inches thick and supported by an aluminum backing ~0.020 inches thick. The activity was sealed in place with another sheet of mylar 0.002 inches thick.

Plots of the efficiencies at several distances are shown in figure 3. With the source close to a large detector, summing reduces the efficiencies for the  $^{88}\text{Y}$  and  $^{60}\text{Co}$  gamma rays in the mixed source. That is, if a radionuclide emits two gamma rays in coincidence, there is a significant probability that both will interact with the detector thus producing the wrong pulse height and not being included in the correct peak area. The dashed lines indicate the expected efficiencies without summing. At larger distances and for smaller detectors, summing was smaller. Note that the shapes of the efficiency curves for the 33% detector are different at low energy because the 33% detector had a beryllium window and the other two had aluminum windows, which attenuated the low energy x-rays, gamma rays, and beta particles. The distances we used were 0 cm with the

80% detector and 1.46 cm with the 33% and 54% detectors. The counting rates at 5.95 cm were too low for our LDEF samples.

### Self Absorption

Self absorption was larger in the LDEF samples than it is in most radiochemistry samples because the LDEF samples were thicker. To determine the self absorption for the 80% detector we placed several different thicknesses ( $x$  in figure 4) of absorber between the source and the detector. The absorber had the same composition as the LDEF samples except, of course, it had not been activated. This procedure varied not only the absorption but also the distance from the source to the detector. Each measurement determined the combined efficiency and self absorption at the distance  $x$  in the LDEF sample. Integrating over  $x$  gives the average combined efficiency and self absorption as a function of the LDEF sample areal density (figure 5).

For the 33% and 54% detectors a different procedure was used because the sample shelf height could only be varied in relatively large steps. We chose to use a shelf height of 1.465 cm. The source mounted on an aluminum planchet was placed on the shelf, and several different thicknesses of absorber were placed on top (figure 6). Representative data and fits are shown in figure 7. The attenuation coefficients are  $\sim 1.2$  to 1.25 larger than those in the literature because of the special geometries.

### Other Factors

Other factors are also involved in quantifying the activation of the samples. Counting statistics were limited by the time the detectors were available. Background determinations were important because radon levels vary and other measurements were in progress in the facilities. Only three sample holders were used with each of the 33% and 54% detectors because we found that backgrounds varied with the sample holders. One holder was found to have appreciable  $^{152}\text{Eu}$  and was not used again. The spatial distributions of activities in the samples can affect the effective efficiencies; we assumed they were uniform. Well known factors include the gamma-ray energies, half-lives, branching ratios, sample masses, and dimensions.

## RESULTS AND DISCUSSIONS

### Trunnion Samples

Figure 8 shows the labeling convention for the trunnion samples. For section D (figure 8b), we analyzed layers two through six on the space side and two through six on the earth side, thus ten layers per trunnion. Since there were two trunnions, east (or right hand) and west (or left hand), we analyzed a total of 20 layers. The top layers, labeled one, which contained  $^7\text{Be}$ , (ref. 2) were analyzed elsewhere. The layers had been flattened when we received them. Note that the thicknesses varied, which complicated the absorption corrections. The material was 17-4 PH stainless steel, which contains about 75% Fe, 15% Cr, 4% Ni, and 3% Cu.

The  $^{54}\text{Mn}$  and  $^{57}\text{Co}$  activities are listed in Table II and plotted in figure 9. Because the procedures and results for the 33% detector and the 54% detector were very similar, the results from these detectors have been combined in column two of Table II. Not all of the samples were counted with the 80% detector because this detector was operated manually and was less available than the others. The uncertainties shown are one standard deviation ( $1\sigma$ ). The values plotted in figure 9 are averages of columns two and three weighted by  $1/s^2$ . Note that the activities near the surface are higher because fewer protons penetrate to the center. The dashed line shows the region for which we did not have samples. Also note that the activities on the west are higher because protons trapped by the earth's magnetic field and striking LDEF on the west side are not limited in energy by the earth's atmosphere. These results are in good agreement with a trapped proton model calculation, (refs. 3 and 4) except near the center where the results are higher, probably reflecting production by galactic cosmic-ray particles. We also analyzed thick 3.25-inch diameter disk samples from sections C, H, and N. Data were taken only with the 80% detector because the samples were too big to fit into the automated systems on the other detectors. Again  $^{54}\text{Mn}$  and  $^{57}\text{Co}$  were detected. Additional studies of the self absorption in these thick samples are required before we can quote reliable values.

Limits on  $^{51}\text{Cr}$ ,  $^7\text{Be}$ ,  $^{22}\text{Na}$ ,  $^{58}\text{Co}$ ,  $^{56}\text{Co}$ ,  $^{46}\text{Sc}$ , and  $^{60}\text{Co}$  were also determined for all of the trunnion samples.

### Aluminum Samples

We analyzed five end support retainer plate samples and two keel plate samples. The material was 6061 aluminum, which contains 1% Mg, 0.6% Si, 0.4% Fe, 0.24% Cu, and 0.2% Cr. Figure 10 shows that  $^{22}\text{Na}$  is clearly present; limits were obtained for  $^7\text{Be}$ . Table III gives the value for  $^{22}\text{Na}$  determined with the 33% and 54% detectors. With the 80% detector  $^{22}\text{Na}$  was seen, but no values are quoted pending more self absorption studies.

## Titanium Samples

Of the nuclides listed above, only  $^{22}\text{Na}$  was detected in the two titanium clips we analyzed (Table IV). The clips used an alloy of titanium with about 6.5% aluminum and 4% vanadium. We only could set limits ( $3\sigma$ ) on the  $^{46}\text{Sc}$ , which should be compared with values for the  $^{54}\text{Mn}$  in the trunnion pieces that is made by a similar nuclear reaction. We detected many gamma-ray lines from uranium and its daughters, which were not expected. We saw lines from  $^{235}\text{U}$  and all of its daughters in equilibrium; we saw lines from the  $^{238}\text{U}$  chain down to  $^{234\text{m}}\text{Pa}$ . Because the same lines have been seen from vanadium, the uranium might have been introduced by the 4% vanadium in this titanium alloy. (ref. 5) These lines will contribute to the background of gamma-ray detectors on spacecraft if this titanium alloy is used nearby.

## CONCLUDING REMARKS

The sensitivity of this experiment was limited not only by the sensitivities of the counting facilities used but also by how soon the samples were available and by the physics. Only a few possible product nuclides emit gamma rays and have sufficiently long half-lives to be counted post flight. Most activation was due to trapped protons, although the galactic cosmic rays contributed significantly to shielded locations. This simple LDEF experiment provided fluence data integrated over a long period of time, which will be useful in designing future spacecraft.

## ACKNOWLEDGMENTS

G. W. Butler provided the 33% and 54% detector systems. L. E. Ussery provided the 80% system. S. G. Bobias assisted in the analysis of the counting results. This work was supported by a contract from NASA/MSFC and done under the auspices of the U.S. Department of Energy.

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TABLE I. MIXED RADIOACTIVE SOURCE

<u>Nuclide</u>	<u>Half-life</u>	<u>Gamma-ray Energies</u> keV
$^{109}\text{Cd}$	464d	88
$^{57}\text{Co}$	272d	122
$^{139}\text{Ce}$	137D	166
$^{203}\text{Hg}$	47d	279
$^{113}\text{Sn}$	115d	392
$^{90}\text{Sr}$	65d	514
$^{137}\text{Cs}$	30y	662
$^{88}\text{Y}$	107d	898,1836
$^{60}\text{Co}$	5.27y	1173,1332



TABLE II. ACTIVITIES IN THE TRUNNION LAYERS

Sample	Activity (picoCuries/kg)			
	$^{54}\text{Mn}$		$^{57}\text{Co}$	
	33% & 54% Detector	80% Detector	33% & 54% Detector	80% Detector
LH, D, Space, 2	$95 \pm 35$	$108 \pm 15$	$27 \pm 17$	$36 \pm 10$
LH, D, Space, 3	$116 \pm 18$		$30 \pm 11$	
LH, D, Space, 4	$86 \pm 17$	$110 \pm 13$	$12 \pm 12$	$8 \pm 9$
LH, D, Space, 5	$89 \pm 16$		$20 \pm 6$	
LH, D, Space, 6	$79 \pm 16$	$99 \pm 10$	$21 \pm 23$	$18 \pm 8$
LH, D, Earth, 2	$145 \pm 40$	$120 \pm 13$	$36 \pm 19$	$20 \pm 8$
LH, D, Earth, 3	$125 \pm 26$		$31 \pm 12$	
LH, D, Earth, 4	$109 \pm 13$	$119 \pm 14$	$19 \pm 10$	$15 \pm 10$
LH, D, Earth, 5	$98 \pm 12$		$12 \pm 8$	
LH, D, Earth, 6	$93 \pm 14$	$99 \pm 12$	$22 \pm 9$	$20 \pm 9$
RH, D, Space, 2	$99 \pm 28$	$104 \pm 21$	$45 \pm 37$	$10 \pm 13$
RH, D, Space, 3	$94 \pm 21$		$-4 \pm 11$	
RH, D, Space, 4	$8 \pm 19$	$81 \pm 11$	$12 \pm 11$	$10 \pm 8$
RH, D, Space, 5	$7 \pm 22$		$35 \pm 22$	
RH, D, Space, 6	$70 \pm 17$	$86 \pm 9$	$-9 \pm 29$	$21 \pm 7$
RH, D, Earth, 2	$116 \pm 31$	$151 \pm 18$	$30 \pm 21$	$13 \pm 11$
RH, D, Earth, 3	$113 \pm 21$		$22 \pm 15$	
RH, D, Earth, 4	$87 \pm 18$	$93 \pm 9$	$12 \pm 11$	$18 \pm 7$
RH, D, Earth, 5	$79 \pm 16$		$10 \pm 15$	
RH, D, Earth, 6	$87 \pm 17$	$71 \pm 10$	$10 \pm 10$	$18 \pm 9$

**TABLE III. ACTIVITIES IN THE ALUMINUM SAMPLES**

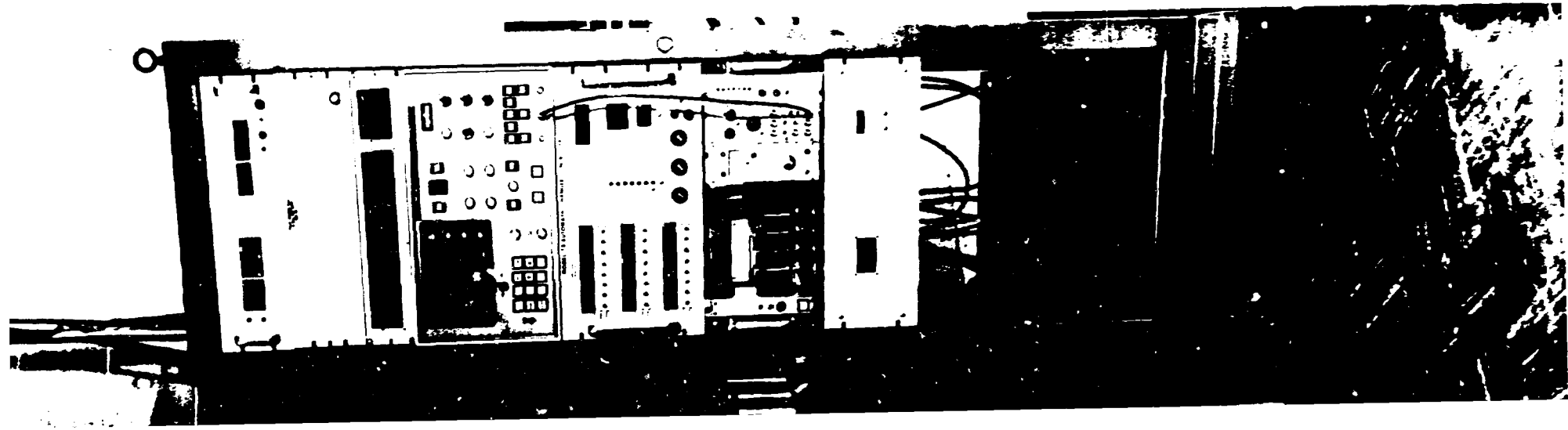
<b>Sample</b>	<b><math>^{22}\text{Na}</math> Activity (picoCuries/kg)</b>
<b>ESR 3</b>	<b><math>103 \pm 17</math></b>
<b>ESR 6</b>	<b><math>113 \pm 19</math></b>
<b>ESR 7</b>	<b><math>114 \pm 25</math></b>
<b>ESR 8</b>	<b><math>122 \pm 29</math></b>
<b>ESR 9</b>	<b><math>117 \pm 18</math></b>
<b>KP 1</b>	<b><math>135 \pm 18</math></b>
<b>KP 12</b>	<b><math>140 \pm 17</math></b>

**TABLE IV. ACTIVITIES IN THE TITANIUM ALLOY SAMPLES**

Sample	<u>Activity (picoCuries/kg)</u>	
	$^{22}\text{Na}$	$^{46}\text{Sc}$
916AC1	$16 \pm 8$	$< 90$
920FC2	$20 \pm 9$	$< 110$

- Figure 1. Automatic counting system used with the 33% detector.
- Figure 2. 80% detector setup.
- Figure 3. Efficiencies measured with the 2" x 2" matrix of sources for the three detectors at various distances.
- Figure 4. 80% detector absorption setup.
- Figure 5. Average efficiency for the 80% detector including the effects of absorption in steel and distance.
- Figure 6. 33% and 54% detector absorption setup.
- Figure 7. Fraction transmitted through the trunnion layers with the 33% or 54% in the setup shown in figure 6.
- Figure 8. LDEF trunnion labeling convention: a) sections and b) layers in section D.
- Figure 9. Induced activity in trunnion section D, a) west and b) east.
- Figure 10. Spectrum for a 4000-minute count of the keel-plate sample KP-12 (top) and a background count of the same length for the same position (bottom) on the 54% detector.





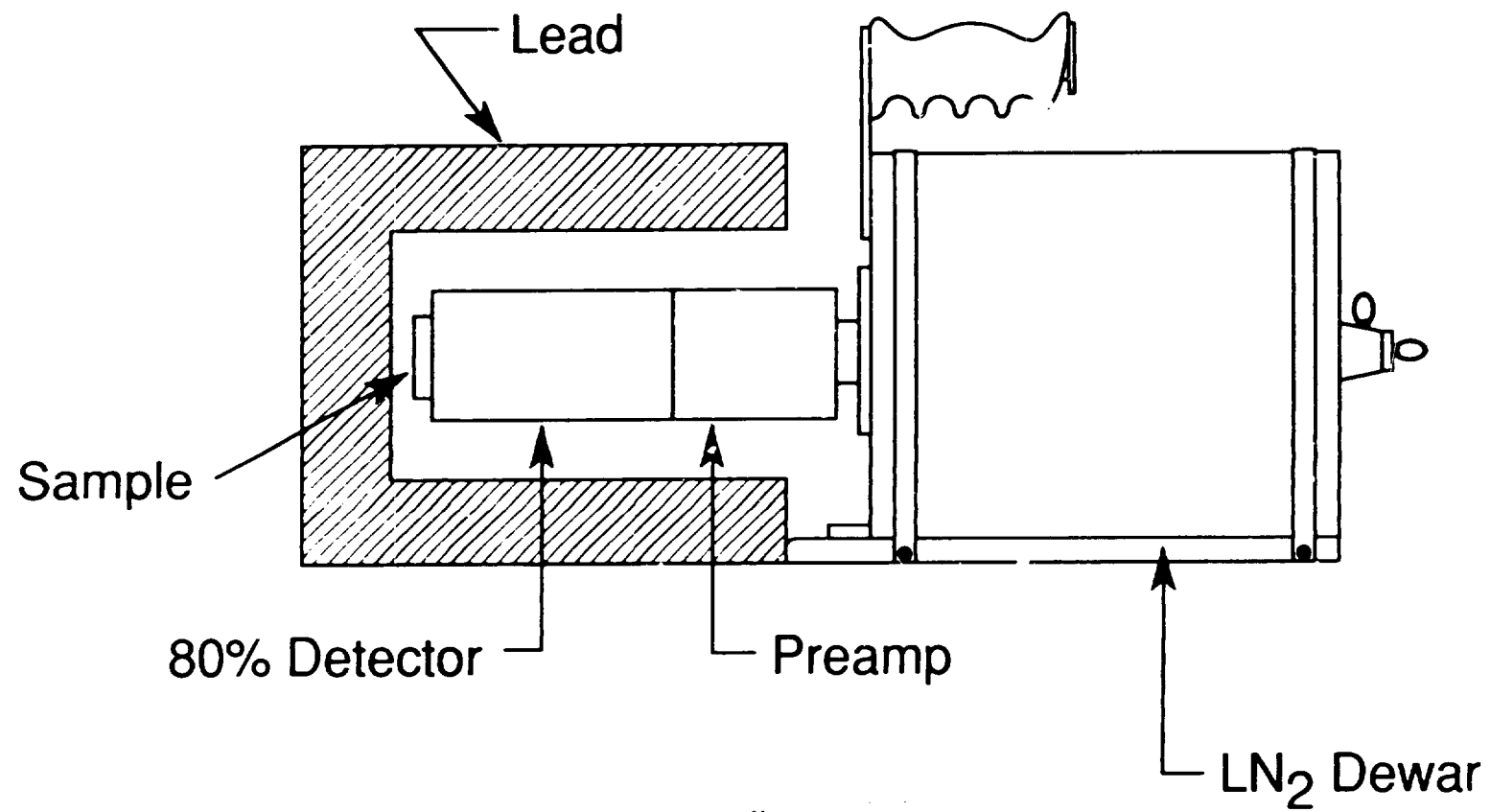


Figure 2

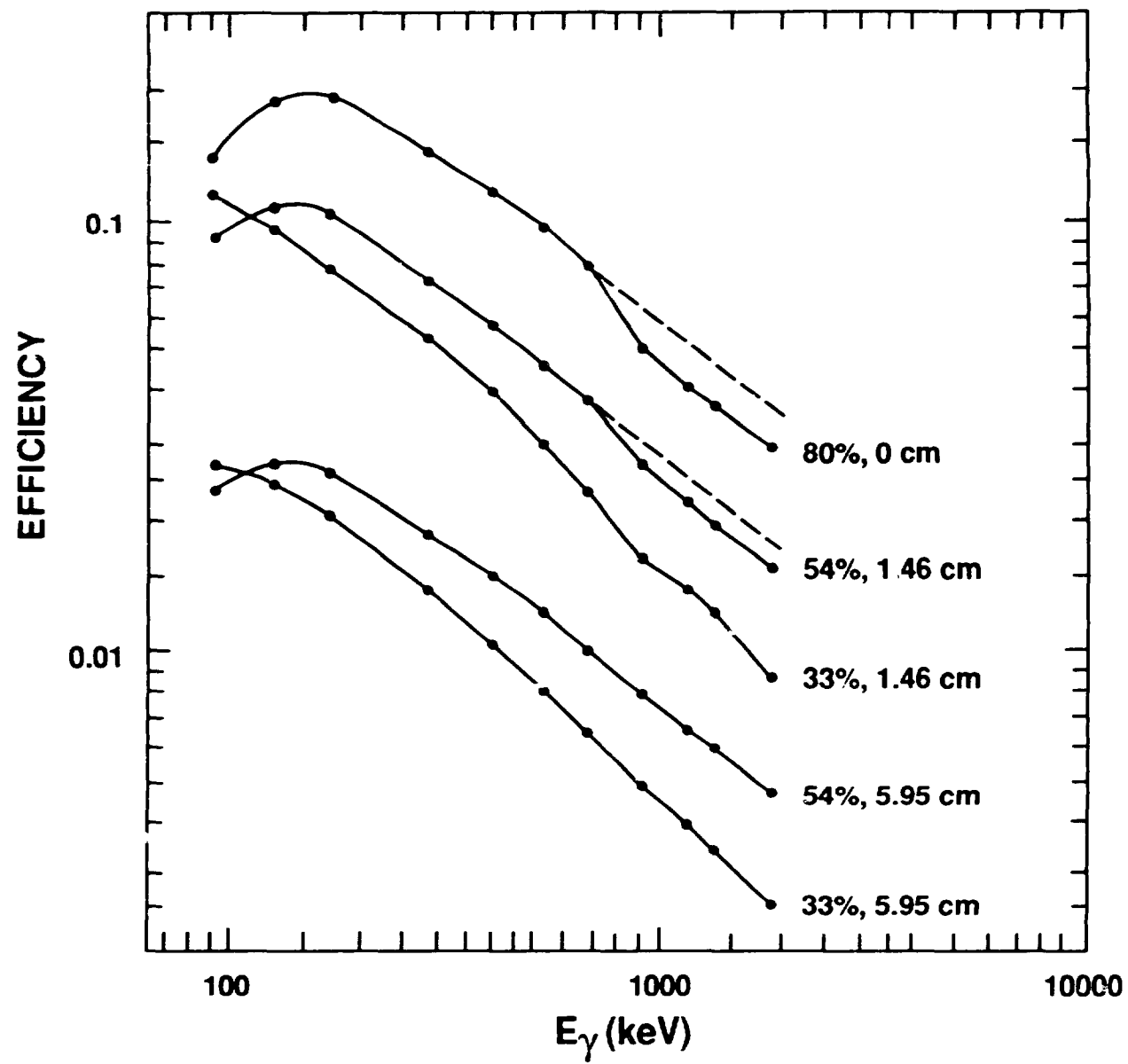


Figure 3



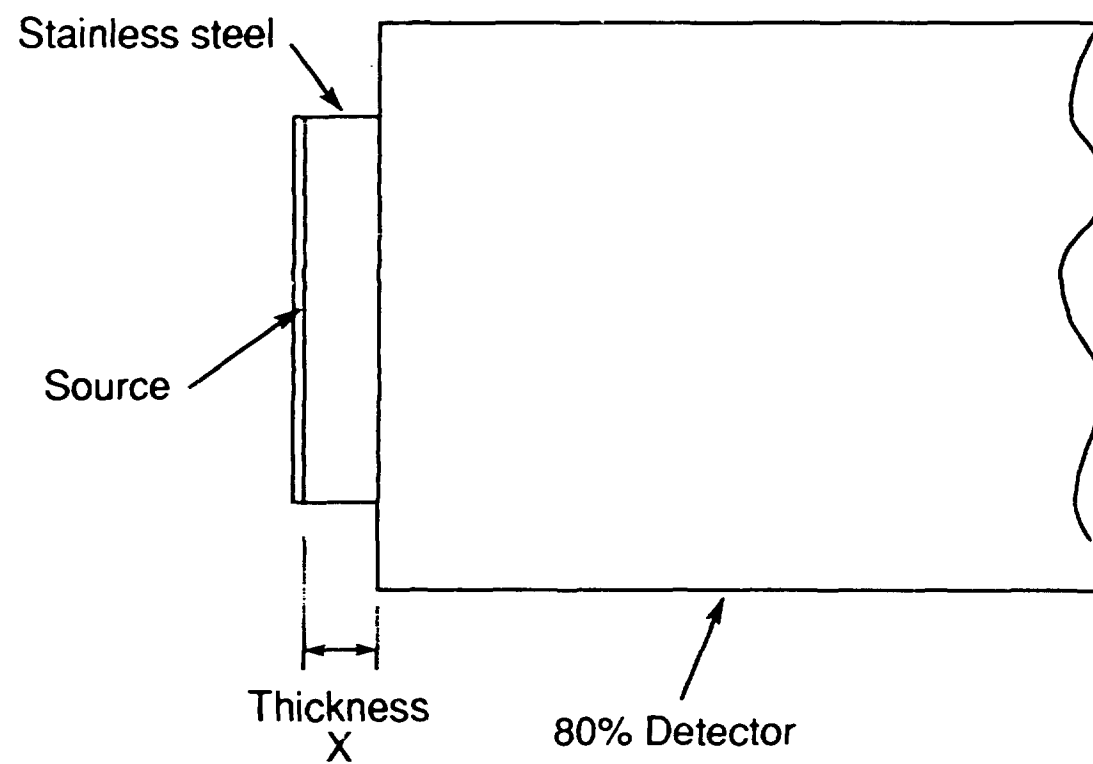


Figure 4

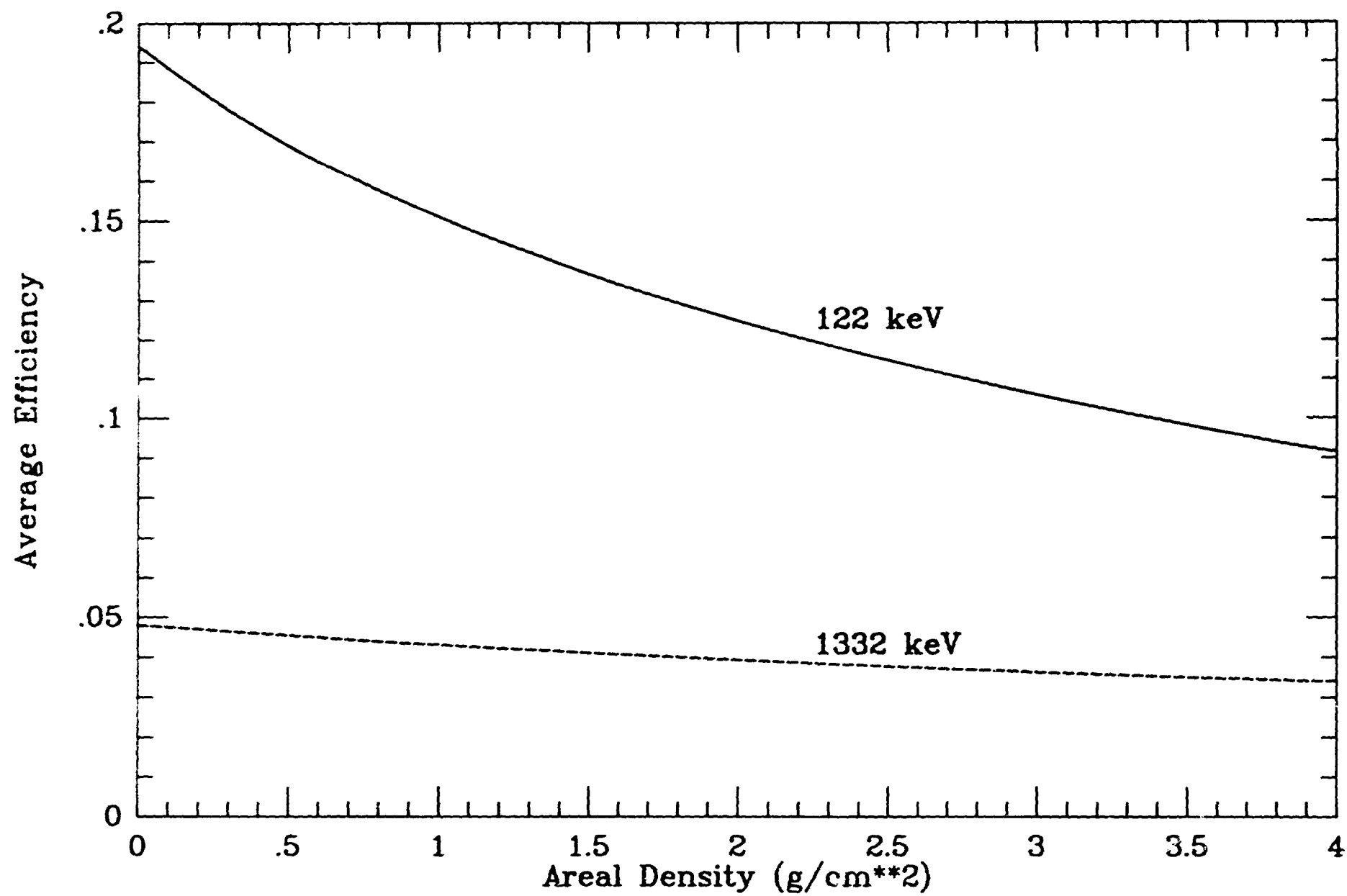


Figure 5

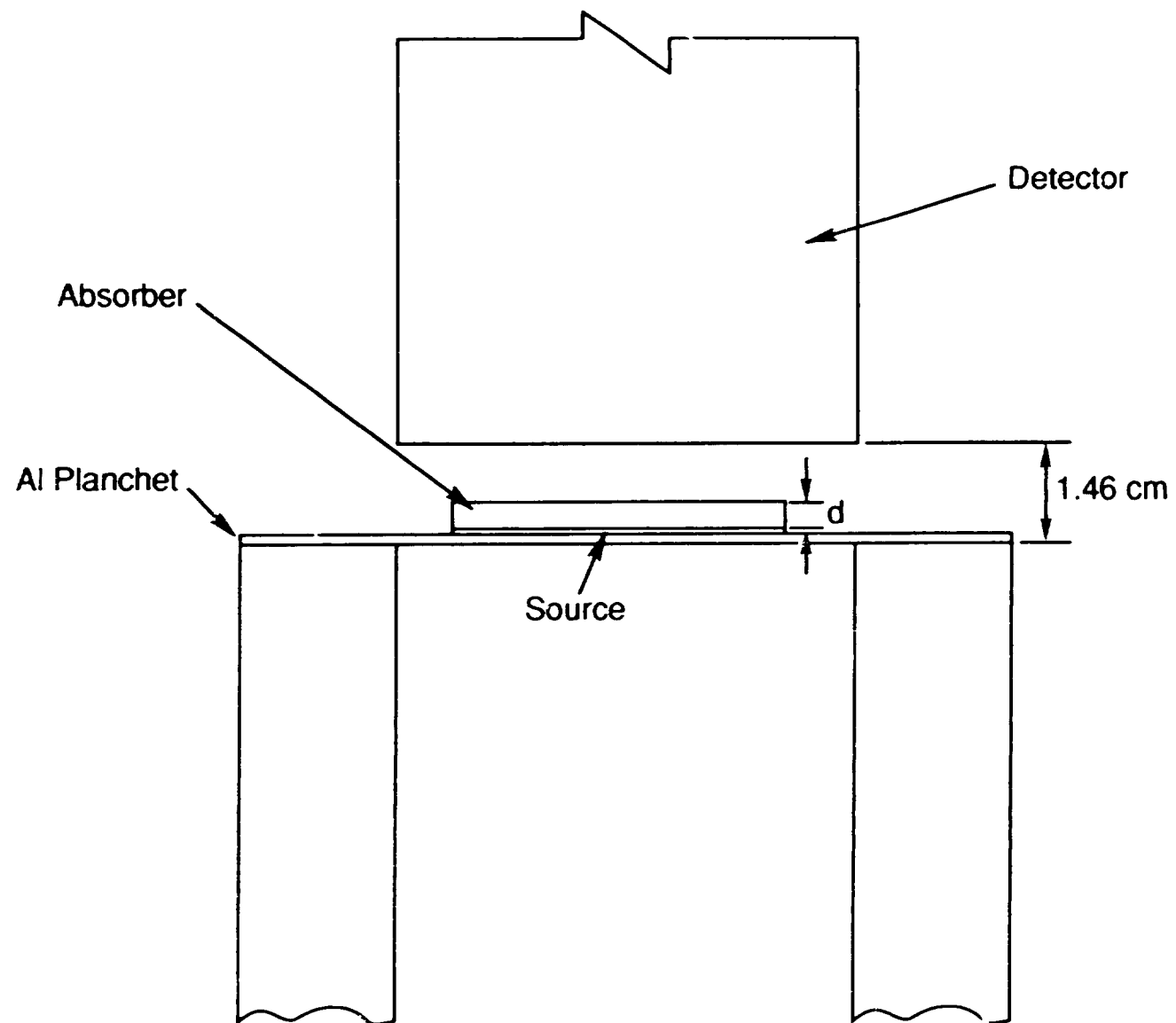
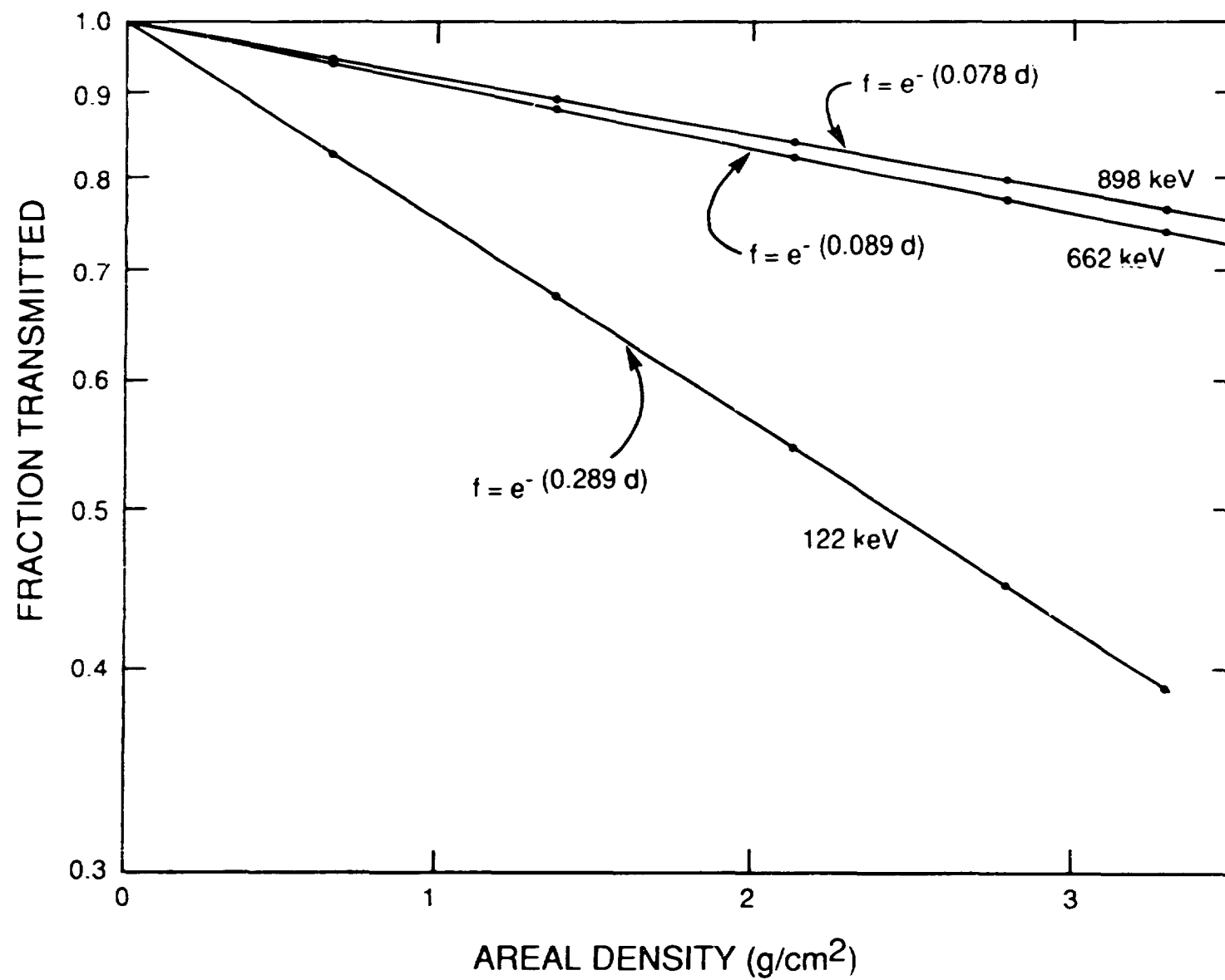


Figure 6



a)

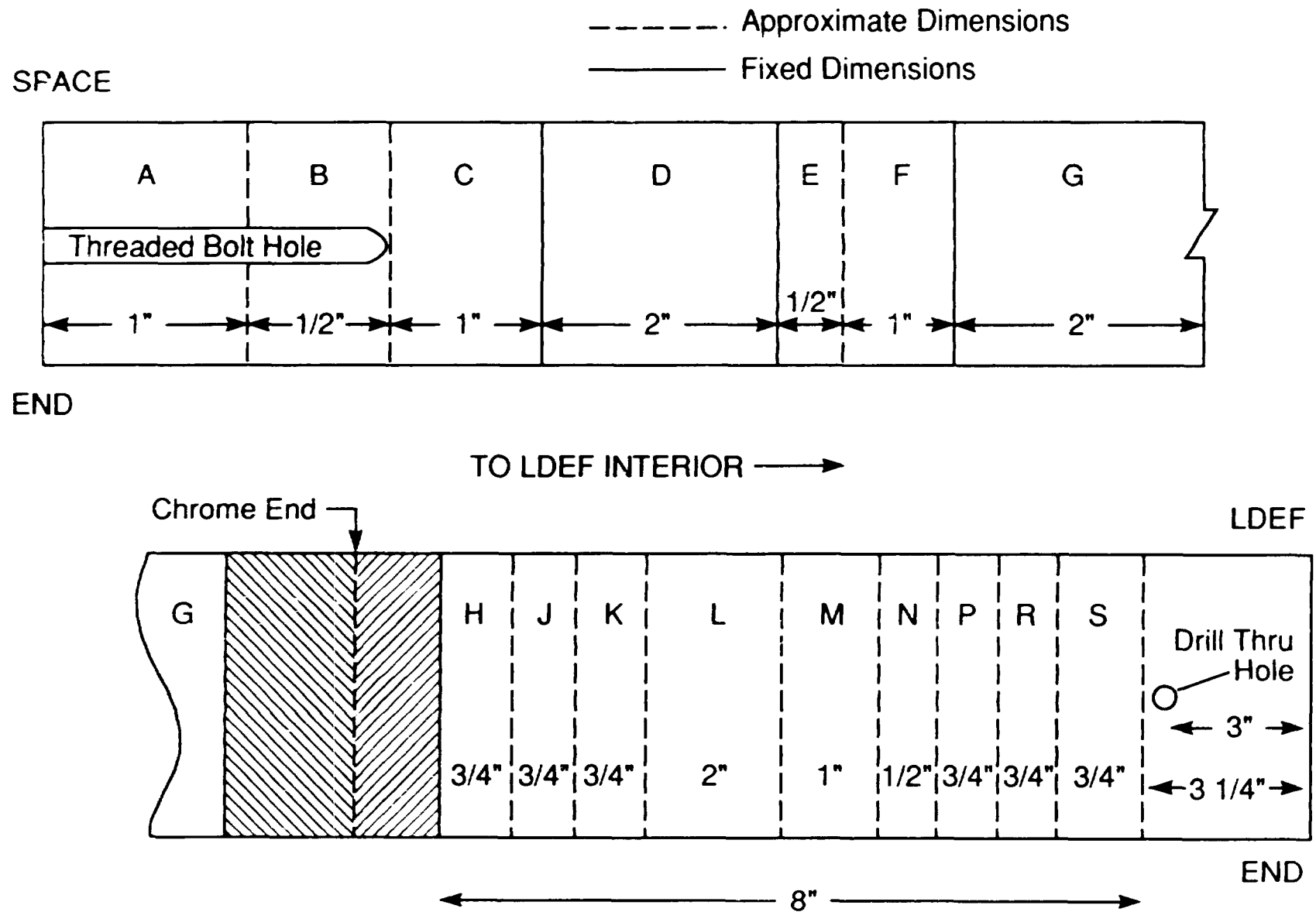


Figure 8a

b)

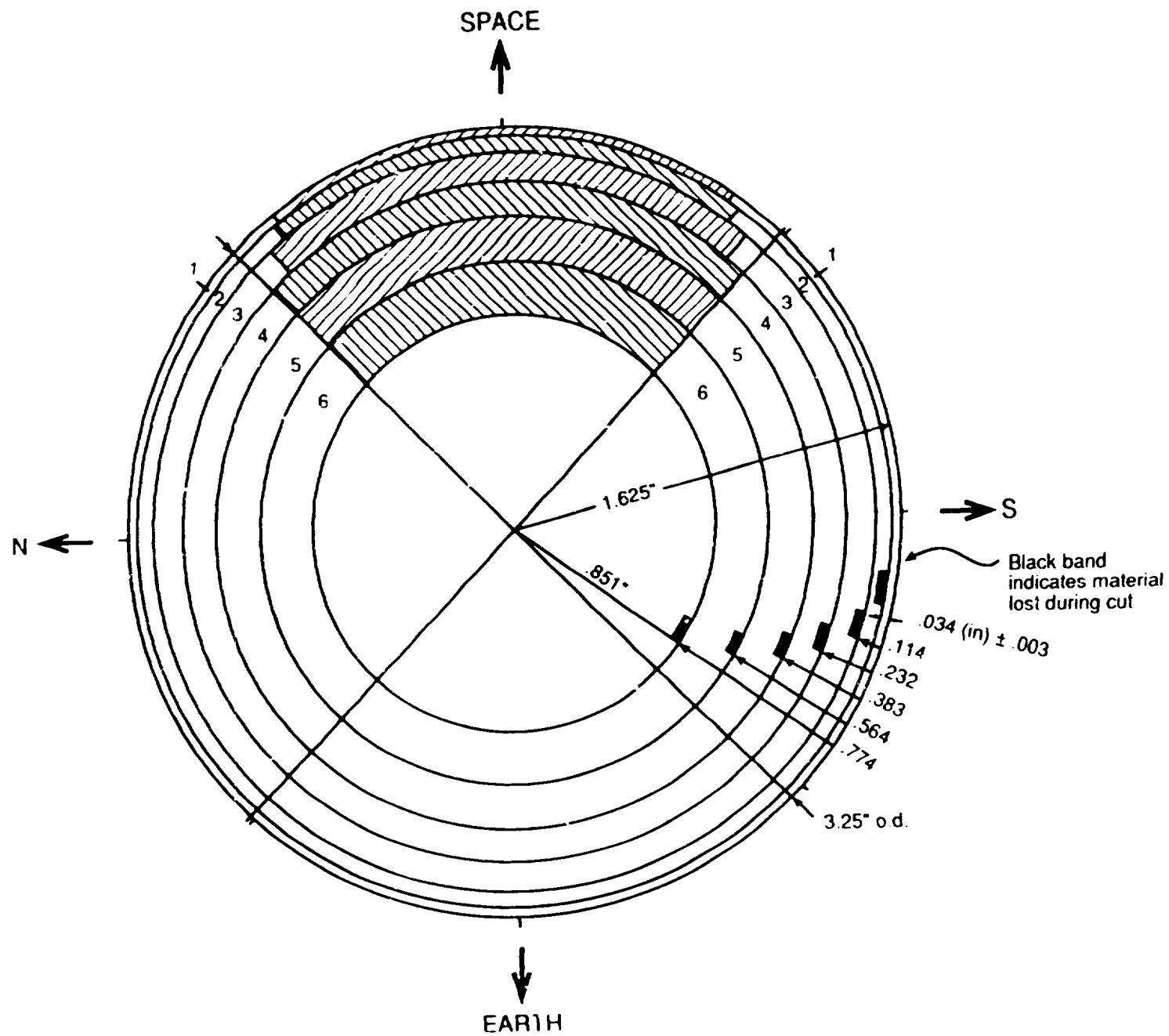


Figure 8b

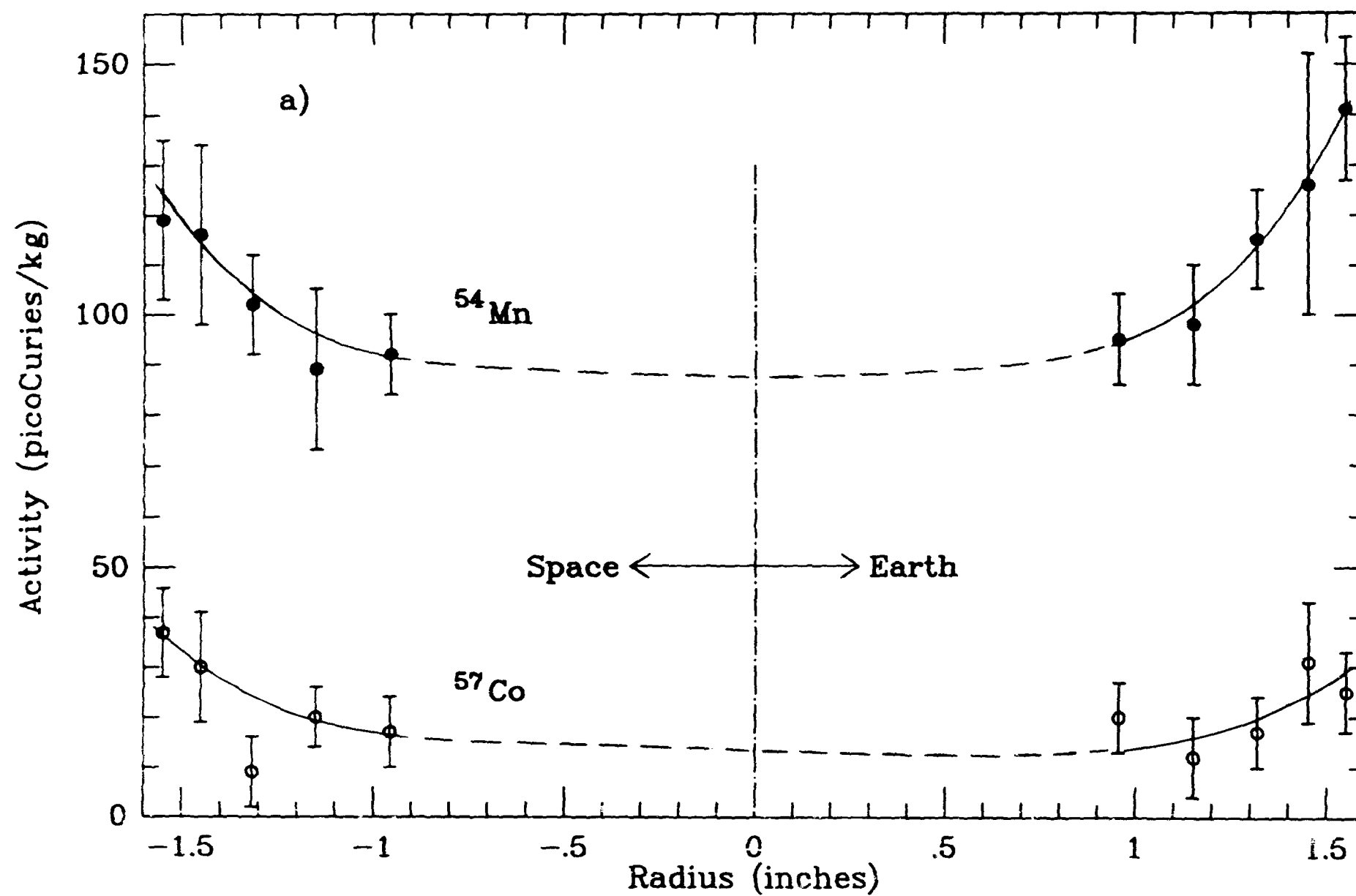


Figure 9a

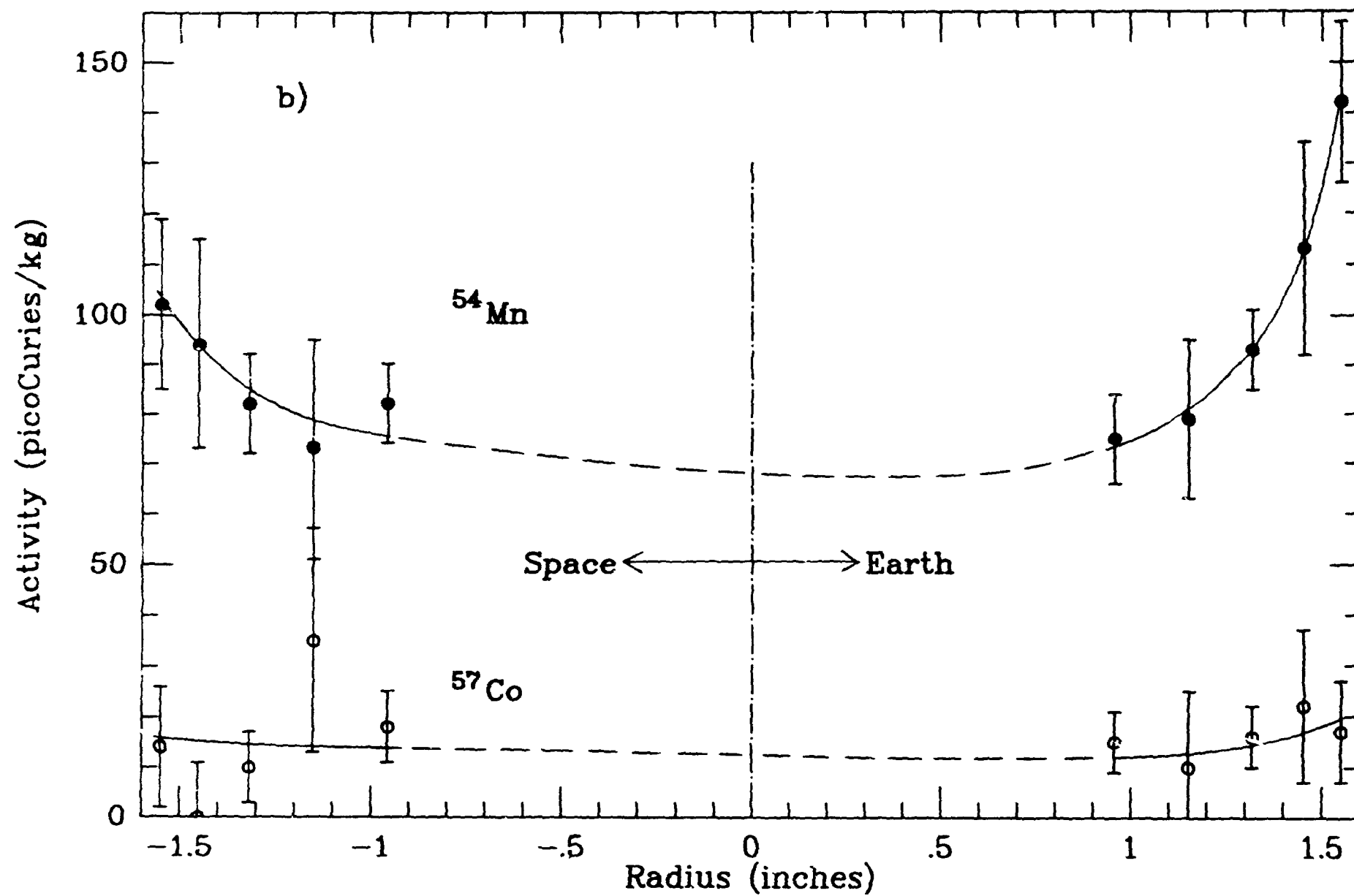


Figure 9b



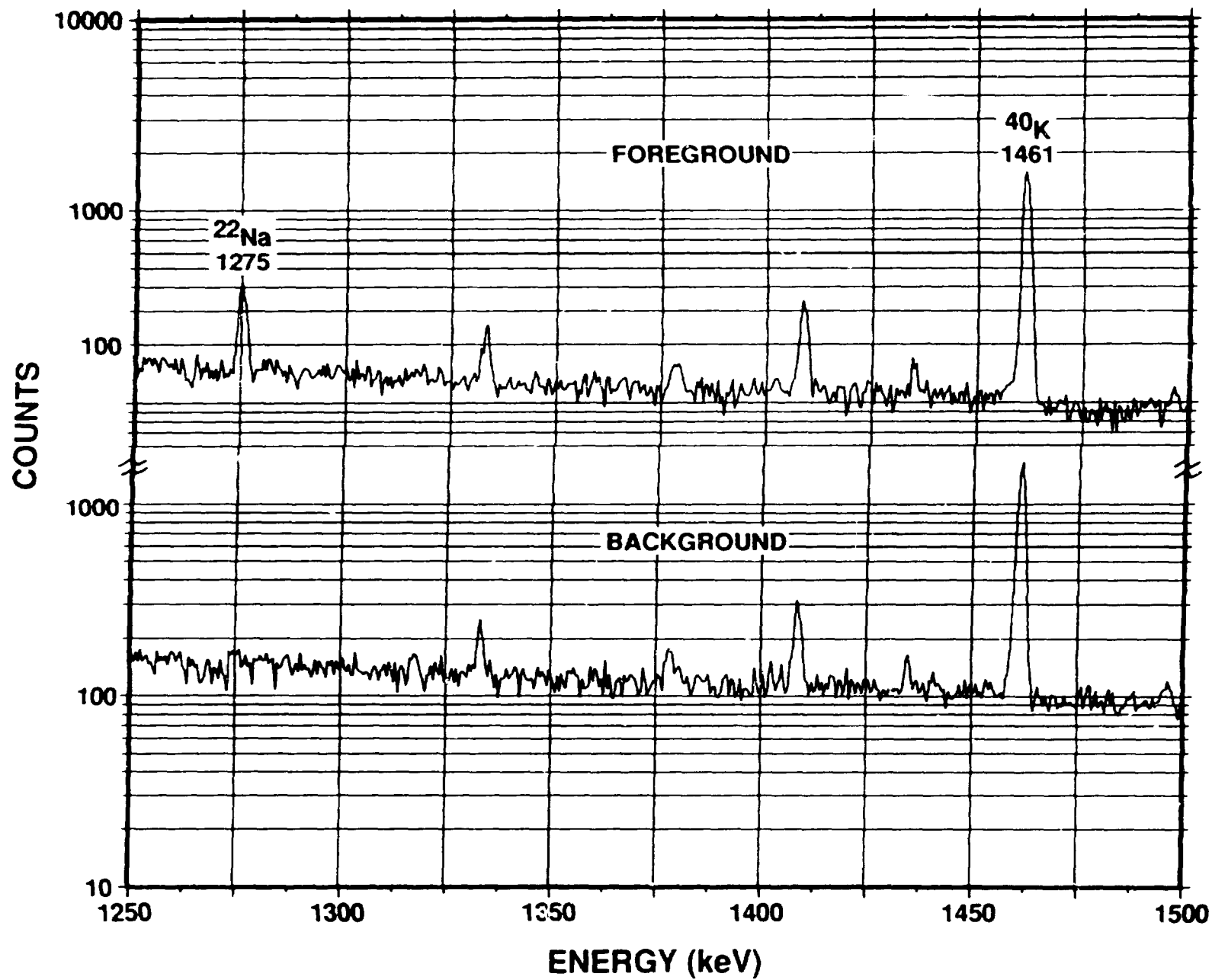


Figure 10